

AMENDMENTS TO THE SPECIFICATION

The paragraph beginning on page 1, line 7, is being amended as follows:

The present invention relates to a functional element for use in an electric, an electronic or an optical device. More particularly, the present invention is concerned with a functional element for use in an electric, electronic or an optical device, comprising a substrate having on an upper surface thereof a plurality of metal oxide needles extending upwardly of the upper surface of the substrate, with their respective central axes arranged substantially in parallel with each other, wherein the metal oxide needles have a specific weighted average circle-based diameter and a specific weighted average aspect ratio, and wherein the metal oxide needles are present at a specific density at the upper surface of the substrate. The present invention is also concerned with a method for producing the above-mentioned functional element. The functional element of the present invention has an advantage in that, although the metal oxide structure therein comprised of the needles has a very large surface area, the metal oxide structure has a very small thickness. Therefore, the functional element of the present invention can be

very advantageously used as a component for an electric, an electronic or an optical device.

The paragraph beginning on page 6, line 18, is being amended as follows:

In this situation, the present inventors have made extensive and intensive studies with a view toward developing a functional element for use in an electric, an electronic or an optical device, which comprises a substrate having a metal oxide structure formed on a surface thereof, wherein the metal oxide structure, on one hand, has a large surface area and, on the other hand, has a small thickness. As a result, the present inventors have successfully developed a functional element, which has been found to have excellent characteristics in that, although the metal oxide structure comprised of the needles has a very small thickness, the metal oxide structure has an extremely large surface area. Specifically, the functional element comprises a substrate having on an upper surface thereof a plurality of metal oxide needles extending upwardly of the upper surface of the substrate, with their respective central axes arranged substantially in parallel with each other, wherein the needles have a weighted ~~an~~ average circle-based diameter of from 0.01 to 10,000  $\mu\text{m}$  and a weighted ~~an~~ average aspect ratio of 0.1 or more and wherein the needles are present at a density of from 0.01 to 10,000 needles per unit area

having a size of  $10\ \mu\text{m} \times 10\ \mu\text{m}$  at the upper surface of the substrate. The present inventors have also found that the above-mentioned functional element can be advantageously applied to various fields including the fields of elements for use in electric or electronic devices, such as an electron emission element of energy saving type (i.e., an electron emission element having the capability of emitting electrons even under low voltages), a high-capacitance capacitor element, a high-density memory element and a high-sensitivity sensor element; and the fields of elements for use in optical devices, such as a laser emission element (particularly a laser emission element emitting a small wavelength laser, such as an ultraviolet laser) and a highly integrated optical switch element. Based on the above findings, the present invention has been completed.

The paragraph beginning on page 10, line 13, and extending to page 11, line 13, is being amended as follows:

In one aspect of the present invention, there is provided a functional element for use in an electric, an electronic or an optical device, comprising a substrate having on an upper surface thereof a plurality of metal oxide needles extending upwardly of the upper surface of the substrate, with their respective central axes arranged substantially in parallel with each other,

wherein the metal oxide needles have a weighted ~~an~~ average circle-based diameter of from 0.01 to 10,000  $\mu\text{m}$ , wherein the weighted average circle-based diameter is defined as the weighted average diameter of circles having areas equal to the areas of the cross-sections of the needles, the cross-sections being taken at the middle portions located at the 1/2 lengths of the needles and along a plane perpendicular to the central axes of the metal oxide needles,

wherein the metal oxide needles have a weighted ~~an~~ average aspect ratio of 0.1 or more, wherein the weighted average aspect ratio is defined as the ratio of the weighted average length of the needles to the weighted average circle-based diameter of the needles, and

wherein the metal oxide needles are present at a density of from 0.01 to 10,000 needles per unit area having a size of 10  $\mu\text{m}$  x 10  $\mu\text{m}$  at the upper surface of the substrate.

The paragraph beginning on page 12, line 12, and extending to page 13, line 9, is being amended as follows:

1. A functional element for use in an electric, an electronic or an optical device, comprising:

a substrate having on an upper surface thereof a plurality of metal oxide needles extending upwardly of the upper surface of the

substrate, with their respective central axes arranged substantially in parallel with each other,

the needles having a weighted ~~an~~ average circle-based diameter of from 0.01 to 10,000  $\mu\text{m}$ , wherein the weighted average circle-based diameter is defined as the weighted average diameter of circles having areas equal to the areas of the cross-sections of the needles, the cross-sections being taken at the middle portions located at the 1/2 lengths of the needles and along a plane perpendicular to the central axes of the needles,

the needles having a weighted ~~an~~ average aspect ratio of 0.1 or more, wherein the weighted average aspect ratio is defined as the ratio of the weighted average length of the needles to the weighted average circle-based diameter of the needles,

the needles being present at a density of from 0.01 to 10,000 needles per unit area having a size of 10  $\mu\text{m}$  x 10  $\mu\text{m}$  at the upper surface of the substrate.

The paragraph beginning on page 16, line 9, is being amended as follows:

The functional element of the present invention comprises a substrate having on an upper surface thereof a plurality of metal oxide needles (i.e., metal oxide whiskers) extending upwardly of the upper surface of the substrate, with their respective central

axes arranged substantially in parallel with each other. The metal oxide needles may have various morphologies, such as a cone-shaped protrusion, a rod, a prism and the like. With respect to the thickness of the metal oxide needles, it is preferred that the metal oxide needles have a weighted ~~an~~ average circle-based diameter of from 0.01 to 10,000  $\mu\text{m}$ , more advantageously from 0.01 to 100  $\mu\text{m}$ , most advantageously from 0.1 to 10  $\mu\text{m}$ . The weighted average circle-based diameter is defined as the weighted average diameter of circles having areas equal to the areas of the cross-sections of the needles, the cross-sections being taken along planes perpendicular to the central axes of the needles, at the middle portions located at the 1/2 lengths of the needles. Specifically, the weighted average circle-based diameter is calculated as follows. The area of the cross-section of a needle is calculated by a conventional method, such as an image analysis. The obtained area of the cross-section is divided by the circular constant  $\pi$ , and, with respect to the resultant value, a square root thereof is obtained. By doubling the obtained square root value, a circle-based diameter of the needle is obtained. Based on the obtained circle-based diameters of the needles, the weighted average circle-based diameter is obtained. When the weighted average circle-based diameter of the metal oxide needles is less than 0.01  $\mu\text{m}$ , it is difficult to stably grow the metal oxide

needles on the surface of the substrate. When the weighted average circle-based diameter is more than 10,000  $\mu\text{m}$ , the desired effect of the surface area increase cannot be satisfactorily achieved by the metal oxide needles.

The paragraph beginning on page 17, line 21, is being amended as follows:

The metal oxide needles have a weighted ~~an~~ average aspect ratio (hereinafter, frequently referred to simply as an "aspect ratio") of 0.1 or more, preferably 0.5 or more, more preferably 1.0 or more, wherein the aspect ratio is defined as the ratio of the weighted average length of the needles to the weighted average circle-based diameter of the needles. When the aspect ratio is less than 0.1, the effect of the surface area increase cannot be achieved by the metal oxide needles. The aspect ratio is preferably 100,000 or less, more preferably 10,000 or less, still more preferably 1,000 or less.

The paragraph beginning on page 18, line 8, is being amended as follows:

With respect to the weighted average length of the metal oxide needles, there is no particular limitation. The desired weighted average length of the metal oxide needles varies depending on the

use of the functional element. However, in general, the weighted average length of the metal oxide needles is preferably from 0.1 to 10,000  $\mu\text{m}$ , more preferably from 1 to 1,000  $\mu\text{m}$ . When the weighted average length is less than 0.1  $\mu\text{m}$ , the effect of the surface area increase cannot be satisfactorily achieved by the metal oxide needles. When the weighted average length is more than 10,000  $\mu\text{m}$ , it is difficult for the functional element to have a satisfactory strength. However, even when the weighted average length is more than 10,000  $\mu\text{m}$ , as described below, a satisfactory strength of the functional element can be achieved by securing the metal oxide needles to each other by means of an organic substance, an inorganic substance or the like.

The paragraph beginning on page 19, line 1, is being amended as follows:

In the present invention, with respect to the metal oxide needles, the weighted average circle-based diameter, the weighted average length and the weighted average aspect ratio are obtained, based on an SEM observation which is made by the following method. First, with respect to a sample of the functional element, the sample is substantially vertically cut along a plane including the center of the upper surface of the sample and extending in parallel to the longitudinal axis of a certain metal oxide needle, to



thereby obtain a cut sample in which a substantially vertical cross-section of the substrate of the functional element is exposed. The cut sample is observed through an SEM, in which the direction of view is toward the cross-section of the substrate. The observation is made with respect to the field of view which extends over 100  $\mu\text{m}$  on both sides of the above-mentioned center of the upper surface of the sample (200  $\mu\text{m}$  in total) as viewed on the cross-section of the substrate. Only the metal oxide needles each exhibiting a completely observable profile (i.e., only the needles each allowing an observation of a complete profile thereof without being obstructed by other needles) are selected. The weighted average circle-based diameter and the weighted average length of the selected metal oxide needles are obtained. The weighted average aspect ratio of the metal oxide needles is obtained as the ratio of the weighted average length of the needles to the weighted average circle-based diameter of the needles.

The paragraph beginning on page 30, line 5, is being amended as follows:

In the production of the functional element of the present invention, for growing a plurality of metal oxide needles on a surface of a substrate, it is required that the application of a metal compound gas onto the surface of the substrate be performed

while keeping the substrate under conditions wherein the substrate is placed in a reaction zone containing an oxide-forming substance and is heated to a temperature higher than the temperature of the metal compound gas. It is preferred that the reaction zone contains air at atmospheric pressure. It is also preferred that the metal compound gas is applied together with a carrier gas which is comprised of an inert gas, such as nitrogen gas. It is required that the contacting of the surface of the substrate with the metal compound gas be performed for a period of time sufficient to grow a plurality of metal oxide needles on the surface of the substrate and form the functional element of the present invention, wherein the functional element comprises a substrate having on an upper surface thereof a plurality of metal oxide needles extending upwardly of the upper surface of the substrate, with their respective central axes arranged substantially in parallel with each other, and must satisfy the features wherein the metal oxide needles have a weighted ~~an~~ average circle-based diameter of from 0.01 to 10,000  $\mu\text{m}$  and a weighted ~~an~~ average aspect ratio of 0.1 or more and wherein the metal oxide needles are present at a density of from 0.01 to 10,000 needles per unit area having a size of 10  $\mu\text{m}$  x 10  $\mu\text{m}$  at the upper surface of the substrate.

The paragraph beginning on page 76, line 9, is being amended as follows:

Representative examples of electric, electronic and optical devices in which the functional element of the present invention can be used are described above. Further examples of such devices to which the functional element is applicable include electric and electronic devices, such as an insulating material, an electroconductive material, a solid electrolyte, a fluorescent image display tube, an EL element, an actuator, a piezoelectric material, a thermistor, a varistor, a superconductive material, a thermoelectric heating/cooling element and an electromagnetic wave shielding material; optical devices, such as a photodielectric material, a light sensor, a solar battery, an optical modulator element and a light absorption filter. The functional element of the present invention has unique features that it comprises a substrate having on an upper surface thereof a plurality of metal oxide needles extending upwardly of the upper surface of the substrate, with their respective central axes arranged substantially in parallel with each other, wherein the metal oxide needles have a large surface area. By virtue of these properties, the functional element of the present invention can also be used in various other devices in addition to the above-mentioned devices. For example, when the metal oxide needles of the functional element

of the present invention have a thickness (weighted average circle-based diameter) of 0.1  $\mu\text{m}$  or less, preferably 0.05  $\mu\text{m}$  or less, the functional element can be used as a thermoelectric heating/cooling element for a refrigerator or the like. Further, recently, studies have been made for using titanium oxide (in combination with a photosensitizer) in a wet type solar battery. In this connection, it should be noted that, when the metal oxide needles of the functional element of the present invention are comprised of titanium oxide, the functional element can be used for a solar battery. In such an application field, since the metal oxide needles in the functional element have a large surface area, a large area can be provided for receiving light energy. Therefore, a solar battery employing the functional element of the present invention exhibits an improved efficiency of the conversion of light into electricity.

The paragraph beginning on page 80, line 18, is being amended as follows:

In order to elucidate the three-dimensional structure of the functional element, SEM photomicrographs showing perspective views of the obtained functional element were taken, and shown in Figs. 2 (a) and 2 (b). The metal oxide ( $\text{ZnO}$ ) needles had a weighted ~~an~~ average circle-based diameter of 1.2  $\mu\text{m}$ , a weighted ~~an~~ average

length of 100  $\mu\text{m}$  and a density of 500 needles per unit area having a size of 10  $\mu\text{m}$  x 10  $\mu\text{m}$ . Further, the leaning angles of the crystal axes of the metal oxide needles were each 0.9 degree.

The paragraph beginning on page 81, line 14, is being amended as follows:

An SEM photomicrograph showing a plan view of the obtained functional element was taken, and shown in Fig. 3. The metal oxide ( $\text{ZnO}$ ) needles had a weighted ~~an~~ average circle-based diameter of 3.6  $\mu\text{m}$ , a weighted ~~an~~ average length of 80  $\mu\text{m}$  and a density of 300 needles per unit area having a size of 10  $\mu\text{m}$  x 10  $\mu\text{m}$ . Further, the leaning angles of the crystal axes of the metal oxide needles were each 0.8 degree.

The paragraph beginning on page 83, line 1, is being amended as follows:

An SEM photomicrograph showing a plan view of the obtained functional element was taken, and shown in Fig. 4. The metal oxide ( $\text{TiO}_2$ ) needles had a weighted ~~an~~ average circle-based diameter of 0.8  $\mu\text{m}$ , a weighted ~~an~~ average length of 5  $\mu\text{m}$  and a density of 2,500 needles per unit area having a size of 10  $\mu\text{m}$  x 10  $\mu\text{m}$ . Further, the leaning angles of the crystal axes of the metal oxide needles were each 2.1 degrees.

The paragraph beginning on page 83, line 18, is being amended as follows:

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In order to examine the three-dimensional structure of the obtained functional element, an SEM photomicrograph showing a perspective view of the functional element was taken, and shown in Fig. 5. The metal oxide ( $\text{TiO}_2$ ) needles had a weighted ~~an~~ average circle-based diameter of  $0.8\ \mu\text{m}$ , a weighted ~~an~~ average length of  $3\ \mu\text{m}$  and a density of 3,200 needles per unit area having a size of  $10\ \mu\text{m} \times 10\ \mu\text{m}$ . Further, the leaning angles of the crystal axes of the metal oxide needles were each  $1.0$  degree.

The paragraph beginning on page 85, line 6, is being amended as follows:

In order to examine the three-dimensional structure of the obtained functional element, an SEM photomicrograph showing a perspective view of the functional element was taken, and shown in Fig. 6. The metal oxide ( $\text{ZnO}$ ) needles had a weighted ~~an~~ average circle-based diameter of  $2.8\ \mu\text{m}$ , a weighted ~~an~~ average length of  $70\ \mu\text{m}$  and a density of 470 needles per unit area having a size of  $10\ \mu\text{m} \times 10\ \mu\text{m}$ . Further, the leaning angles of the crystal axes of the metal oxide needles were each  $3.9$  degrees.

The paragraph beginning on page 86, line 20, is being amended as follows:

In order to examine the three-dimensional structure of the functional element, an SEM photomicrograph showing a perspective view of the obtained functional element was taken, and shown in Fig. 7. The metal oxide (ZnO) needles had a weighted ~~an~~ average circle-based diameter of 0.25  $\mu\text{m}$ , a weighted ~~an~~ average length of 0.5  $\mu\text{m}$  and a density of 2,000 needles per unit area having a size of 10  $\mu\text{m}$  x 10  $\mu\text{m}$ .